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(54) Title: BINDERS FOR COMPOSITE PANELS

(57) Abstract: A binder for composite panels of the type including particleboard, fibreboard, plywood and other products made from combinations of particles, fibres, wafers, strands and veneers. The binder is formed by the reaction of formaldehyde with urea and/or melamine, to achieve a molar ratio which will result in a formaldehyde emission of a desired level. Isocyanate is added to reverse loss in physical and mechanical properties arising from use of the binder in the composite panel.

TITLE OF THE INVENTION

BINDERS FOR COMPOSITE PANELS

BACKGROUND OF THE INVENTION

This invention relates to improvements in and relating to binders and more particularly improved binders for use in the manufacture of composite panels.

Binders are used in the production of composite panels and plywood. The binders can be made by reacting formaldehyde with urea and/or melamine, referred to herein as simply "(M)UF binders". The formaldehyde reacts with amine groups in the urea and/or melamine. The formaldehyde acts as a crosslink to form polymers.

The binders are used to make composite panels from lignocellulosic materials such as wood, bagasse, straw and agricultural residues. Composite panels may include particleboard, fibreboards including medium density fibreboard (MDF), waferboard, strandboard including oriented strandboard (OSB), plywood and other products made from combinations of particles, fibres, wafers, strands and veneers. Throughout the following disclosure the reference to "composite panels" will include such boards, plywood and other products.

Composite panels made using a binder of the type to which this invention relates commonly emit gaseous

formaldehyde. The formaldehyde emissions are related to the quantity of formaldehyde used to manufacture the binder. The quantity of formaldehyde in the binder may be expressed relative to the amount of amine functional groups as the formaldehyde-amine molar ratio ($F:NH_2$). In general, the lower the $F:NH_2$ ratio the lower the quantity of formaldehyde which is emitted

There are a number of recognised methods of measuring the emission of formaldehyde. One method is to extract the formaldehyde with toluene. The total extractable formaldehyde is related back to the mass of the composite panel. The amount of formaldehyde expressed as mg/100g of bone dry composite panel is commonly referred to as the perforator value. The test method is described in the CEN Standard EN120.

Other test methods/standards include J1S A5905-1994 Fibreboards and J1S A5908-1994 Particleboard, F1 formaldehyde emission as measured by JAS Structural Plywood and LVL Standards.

These methods measure the amount of formaldehyde emitted from pieces of composite panel placed in a desiccator. The emitted formaldehyde is absorbed into water and expressed as mg/litre of water. The methods are commonly referred to as the Japanese desiccator method.

In accordance with the present invention, a formaldehyde emission less than or equal to EO as measured by the Japanese desiccator method or the CEN Standard EN120 equivalent perforator value is the objective.

The $F:NH_2$ ratio of the binders used in the construction of composite panels influences some of the physical and mechanical properties of the composite panel. By way of example, the tensile stress perpendicular to the plane of the panel (internal bond) and the thickness swell caused by immersion of the composite panel in water typically deteriorates as the $F:NH_2$ ratio decreases. Therefore, when making composite panels which have very low emissions of formaldehyde (ie less than or equal to EO) the $F:NH_2$ ratio is often so low that the binder is unable to impart the desired physical and mechanical properties to the composite panel.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide a binder which can be used to make composite panels with ultra-low formaldehyde emissions yet with desired physical and mechanical properties.

An object of the present invention is thus the production of composite panels having ultra-low formaldehyde emissions.

According to one broad aspect the present invention provides a binder for composite panels as herein defined, the binder being formed by reaction of formaldehyde with urea and/or melamine to achieve a molar ratio which will result in a formaldehyde emission in a composite panel of the desired level and adding an isocyanate to reverse loss in physical and mechanical properties arising from use of the binder in the composite panel.

A second broad aspect of the invention provides a method of producing a composite panel as herein defined, the method being characterised by using a binder of the first broad aspect to bind the material of at least a core layer of the composite panel.

According to a third broad aspect there is provided by the invention a composite panel as herein defined made using a binder method of the type set forth in the first or second broad aspect.

In the preferred form the molar ratio is selected to result in a formaldehyde emission of equal to or less than EO.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present invention in a preferred form is based on the selection of a binder with a $F:NH_2$ ratio which gives the desired ultra-low formaldehyde emission. To this binder

is added a quantity of the isocyanate commonly known as MDI or PMDI. The resultant (M)UF/MDI blend provides a binder for composite panels made of lignocellulosic materials which exhibits the desired end use physical and mechanical properties. This is achieved due to the MDI component reversing the losses in the physical and mechanical properties arising out of the use of the selected (M)UF binder.

The quantity of (M)UF binder, expressed as the percentage of binder solids based on the bone dry mass of the lignocellulosic material, may be between substantially 1 and 20% depending on the nature of the composite panel and the desired physical and mechanical properties. More commonly, the quantity of (M)UF binder is between 3 and 18%. Preferably, the amount used is between substantially 5 and 15%.

The quantity of MDI added depends on the desired physical and mechanical properties and the desired level of formaldehyde emission. With the objective of meeting formaldehyde emission less than or equal to EO, the quantity of MDI added will typically be between substantially 0.1 and 4% of the bone dry mass of lignocellulosic material. More commonly, it is between 0.3 and 3% preferably between substantially 0.5 and 2%.

Furthermore, the MDI which is added may be used to substitute for some or all of the melamine component of

(M)UF binders without incurring a significant decline in the moisture resistant properties of the composite panels.

To more fully describe the invention reference will now be made to the following examples.

Example 1

Laboratory-scale composite panels with surface layers made using fibre containing an MUF binder alone and a core layer made with randomly oriented strands of nominal size 75x5x0.5mm using different combinations of the MUF binder and MDI were evaluated for formaldehyde emissions and thickness swell following immersion in water. A formaldehyde scavenger containing urea was incorporated as part of the MUF binder to give different $F:NH_2$ ratios. The composite panels were 30mm thick after sanding and the nominal density was $700g/m^3$.

The fibre for the surface layers was blended with 15% of MUF binder. The strands for the core layer were coated with combinations of MUF binder and MDI as noted in Table 1. Some physical properties including formaldehyde emissions are included in the table.

The properties are summarised in Table 1.

Table 1

Treatment No	1	2	3	4
Strand binder				
MUF (%)	10	10	10	10
Scavenger (%)	0	0	0.1	0.2
$F:NH_2$	0.43	0.43	0.425	0.42
MDI (%)	0	1	1	1
Formaldehyde emission(mg litre)	0.62	0.73	0.48	0.41
Thickness swell(20°C.24h)	11	7	8	10
Thickness swell (100°C, 10min)	68	59	46	55

Example 2

Composite panels 30mm thick were manufactured in the laboratory. The surface layers of the composite panels were made from wood fibre containing an MUF binder alone. The core layer of the composite panels consisted of randomly oriented wood strands of nominal size 75x5x0.5mm blended with different combinations of MUF binders and MDI. The melamine content of the MUF binders used in the core layer varied between 3 and 30%. A formaldehyde scavenger containing urea was incorporated as part of the core layer MUF binder to give different $F:NH_2$ ratios. The composite panels were 30mm thick after sanding and the nominal density was 700kg/m^3.

The fibre for the surface layers was blended with 15% of MUF binder. The strands for the core layer were coated with combinations of MUF binder and MDI as noted in Table

2. Some physical properties including formaldehyde emissions are included in the table.

Table 2

Treatment No	1	2	3	4	5
Strand binder					
MUF(%)	10	10	10	10	10
Scavenger (%)	0	0	0.1	0.2	0.2
$F:NH_2$	0.43	0.43	0.425	0.42	0.42
Melamine content (%)	30	30	30	10	3
MDI (%)	0	1	1	1	1
Formaldehyde emission (mg/litre)	0.95	0.81	0.60	0.54	0.51
Thickness swell (20°C, 24h)	10	9	8	7	5
Thickness swell (70°C, 2h)	28	23	22	31	24

Example 3

A series of composite panels were manufactured in a composite panel plant. The composite panels had surface layers made using wood fibre containing an MUF binder and a core layer made from randomly oriented wood strands of nominal size 75x5x0.5mm using different combinations of MUF resins and MDI. A formaldehyde scavenger containing urea was incorporated as part of the core layer MUF binder to give different $F:NH_2$ ratios. The thickness of the composite panels after sanding was 30mm and the nominal density was 600kg/m^3.

The fibre for the surface layers was blended with 15% of the MUF binder in the blowline. The strands for the core layer were coated with combinations of (M)UF binders and MDI in a rotary drum blender as noted in Table 3. The internal bonds of the composite panels and their swelling characteristics in water under various conditions were measured (see Table 3).

Table 3

Treatment No	1	2	3	4	5	6
Strand binder						
MUF (%)	10	10	10	10	10	10
Scavenger (%)	0	2	2	2	1	1
$F:NH_2$	0.43	0.34	0.34	0.34	0.38	0.38
Melamine content(%)	30	10	10	30	30	10
MDI (%)	0	1	2	2	2	2
Formaldehyde emission (mg/litre)	0.82	0.24	0.21	0.26	0.39	0.34
Internal bond (kgf/cm^2)	0.36	0.16	0.26	0.37	0.41	0.41
Thickness swell (20°C, 24h)	4.7	7.7	5.8	3.9	4.2	4.9

Example 4

A series of composite panels was manufactured in the laboratory using (M)UF binders containing from 0 to 50% melamine based on solids and MDI. The surface layers of

the composite panels consisted of wood fibres with an (M)UF alone as the binder. The core layer of the composite panels consisted of wood strands of nominal size 75x5x0.5mm, randomly oriented and with various combinations of (M)UF binder and MDI.

The fibre for the surface layers was blended in a laboratory blender with 15% of an MUF binder. The strands for the core layer were coated with combinations of (M)UF and MDI as noted in Table 4. The internal bonds of the composite panels and their swelling characteristics under various conditions were measured (see Table 4).

Table 4

Treatment No	1	2	3	4	5	6	7	8
Strand binder								
MUF (%)	15	10	10	10	10	10	10	15
Melamine content(%)	30	0	0	3	13	30	50	30
MDI (%)	0	0	1	1	1	1	1	0
Internal bond (kg/cm^2)	1.23	0.77	0.99	1.00	1.12	1.03	1.10	1.24
Thickness swell (20°C,24h)	9.6	13.6	10.4	8.1	9.6	8.8	8.3	10.6
Thickness swell (70°C,2h)	18.8	37.3	21.9	28.6	18.9	17.3	16.6	18.2
Thickness swell (100°C,10min)	16.0	31.7	9.7	16.0	11.1	14.2	14.3	14.1

The present invention thus provides a melamine-urea-formaldehyde/diphenylmethane-di-isocyanate (MUF/MDI) blend as a binder for composite panels made from

lignocellulosic materials with a formaldehyde emission level of less than EO.

The use of PMDI may lead to a reduction in the amount of melamine in the (M)UF resin component without loss of moisture resistance properties. It is possible to obtain thickness swells better than those obtained with a 50% melamine resin BOS with a resin containing less than 50% melamine.

It is also possible to use an (M)UF resin and PMDI to improve the thickness swells of composite panels.

The PMDI addition compensates for the loss of product properties brought about by the low quantities of formaldehyde in the (M)UF resins.

(M)UF may contain an added quantity of UF resin and/or urea solution and/or other formaldehyde scavenger with a preferred option being an (M)UF resin.

By use of the binder according to the present invention, production of composite panels having ultra-low formaldehyde emissions (emissions less than or equal to EO) can be achieved without adversely impacting on the physical and mechanical properties of the composite panel.

CLAIMS

1. A binder for composite panels as herein defined, the binder being formed by reaction of formaldehyde with urea and/or melamine to achieve a molar ratio which will result in a formaldehyde emission in a composite panel of the desired level and adding an isocyanate to reverse loss in physical and mechanical properties arising from use of the binder in the composite panel.

2. The binder as claimed in claim 1 wherein the molar ratio is selected to result in a formaldehyde emission of equal to or less than EO.

3. The binder as claimed in claim 1 or 2 wherein the isocyanate is diphenylmethane-di-isocyanate.

4. A method of producing composite panel as herein defined, the method characterised by using a binder according to claim 1, 2 or 3 to bind the material of at least a core layer of the composite panel.

5. The method as claimed in claim 4 wherein the quantity of binder solids based on the bone dry mass of the lignocellulosic material is between substantially 1% and 20%.

6. The method as claimed in claim 5 wherein the quantity of binder is between substantially 3% and 18%.

7. The method as claimed in claim 5 wherein the quantity of binder is between substantially 5% and 15%.

8. The method as claimed in any one of claims 4 to 7 wherein the quantity of isocyanate is between substantially 0.1% and 4% of the bone dry mass of lignocellulosic material.

9. The method as claimed in claim 8 wherein the quantity of isocyanate is between substantially 0.3% and 3%.

10. The method as claimed in claim 8 wherein the quantity of isocylanate is between substantially 0.5% and 2%.

11. A composite panel produced according to the method of any one of claims 4 to 10.

12. A composite panel produced by using a binder according to any one of claims 1 to 3 to bind the material of at least a core layer of the composite material.

13. A binder for composite panels as herein defined as claimed in claim 1 substantially as herein described with reference to the accompanying Examples.

14. A composite panel when produced by the method according to claim 4 substantially as herein described with reference to the accompanying Examples.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/NZ00/00236

A.	**CLASSIFICATION OF SUBJECT MATTER**		
Int. Cl. [7]:	C08G 18/54 C08L 97/02, 75/12 C09J 175/12		
According to International Patent Classification (IPC) or to both national classification and IPC			

B.	**FIELDS SEARCHED**
Minimum documentation searched (classification system followed by classification symbols)	
C08G 18/54 C08L 97/02, 75/12 C09J 175/12	
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched	
AU: IPC AS ABOVE	
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)	
WPAT: +ISOCYANAT+ and FORMALDEHYD+ and (UREA+ or MELAMIN+) and IPC as above JAPIO: as above for WPAT	

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	AU 11424/00 A (ORICA AUSTRALIA PTY. LTD.) 4 May 2000 Examples 1-5	1-14
X	FR 2592382 A (SOCIETE CHIMIQUE DES CHARBONNAGES S.A.) 3 July 1987 Examples 1-4	1,4-12
X	EP 107260 A (METHANOL CHIMIE NEDERLAND V.O.F. and STAMICARBON B.V.) 2 May 1984 Examples I, III and IV	1-14

☒ Further documents are listed in the continuation of Box C ☒ See patent family annex

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Date of the actual completion of the international search	Date of mailing of the international search report
5 February 2001	8 February 2001
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA E-mail address: pct@ipaustralia.gov.au Facsimile No. (02) 6285 3929	Authorized officer N.L. KING Telephone No : (02) 6283 2150

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4362827 A (Tinkelenberg et al.) 7 December 1982 Examples I, II, IV, V	1-14
A	JP 59179573 A (NIPPON URETHANE SER) 12 October 1984) Derwent Abstract No. 84-291409 Abstract	1-14
A	JP 58157876 A (MITSUI TOATSU CHEM INC) 20 September 1983 Derwent Abstract No. 83-798762 Abstract	1-14
A	SU 804513 A (DNAPR CHEM TECH INST) 15 December 1981 Derwent Abstract No. 81-83185D Abstract	1-14

INTERNATIONAL SEARCH REPORT Information on patent family members	International application No. **PCT/NZ00/00236**

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report		Patent Family Member					
AU	11424/00	WO	200024800				
FR	2592382	EP	232642	AT	85061	DE	3687652
		ES	2053453				
EP	107260	NL	8204144	NO	833915		
US	4362827	CA	1166394	DK	3771/80	EP	25245
		FI	802829	NL	7906751		

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